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Summary of thesis: Self-propelled motion of a fluid droplet under chemical reaction

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Self-propelled motion of particles has attracted much attention from the viewpoint of non-linear physics far from equilibrium. There are several experiments of self-propulsion of a fluid droplet [1, 2] and theoretical studies of droplet motion due to interfacial tension gradient along the droplet surface [3]. However, these theories are concerned only with the steady velocity of a droplet.

It should be noted that self-propelled motion of particles has been investigated in a different field of physics. Numerical simulations of a reaction-diffusion equation indicated that changing system parameters induces a bifurcation from a motionless state to a propagating state [4]. The domain dynamics near the drift bifurcation in a reaction-diffusion system was investigated theoretically [5].

We studied self-propelled dynamics due to the coupling between a Marangoni effect and chemical reaction by extending the previous studies on the reaction-diffusion systems [6]. We introduced a model system of phase separating binary mixture where a chemical reaction takes place inside a droplet. The free energy of this model is given in terms of $\phi = \phi_A - \phi_B$ as

$$F\{\phi\} = \int d\vec{r} \left[\frac{B(c)}{2} (\nabla\phi)^2 + f(\phi) + k_B T n c \ln c \right], \tag{1}$$

where $\phi_{A(B)}$ is the local concentration of the component A(B). n is the molecular number density of the system. c is the density of the third dilute component. $f(\phi)$ is a free energy density such that phase separation takes place at low temperatures. The surface tension $\sigma = \sigma_0 + c\sigma_1 > 0$ is assumed to depend linearly on c. The time-evolution equations for ϕ and c are given by

$$\frac{\partial \phi}{\partial t} + \nabla \cdot (\vec{v}\phi) = L\nabla^2 \frac{\delta F}{\delta \phi}, \tag{2}$$

$$\frac{\partial c}{\partial t} + \nabla \cdot (\vec{v}c) = D\nabla^2 c - \gamma c + A\theta \left(R - |\vec{r} - \vec{r}_G| \right), \tag{3}$$

The third component is produced inside the droplet and diffuses away from the droplet. The coefficient A stands for the rate of the production of the third component inside the droplet.

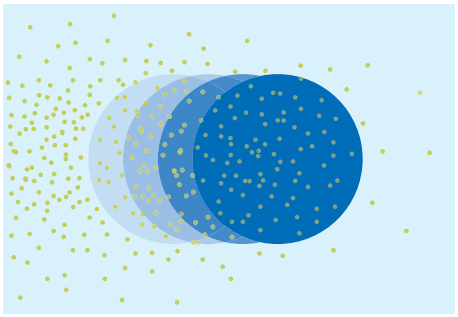

Figure 1: Translational motion of a droplet. The droplet is migrating to the right under the non-uniform distribution of the c component indicated by the small circles.

The Stokes approximation is employed for the local velocity $\vec{v}$ and it takes the form

$$0 = -\vec{\nabla}p - \phi\vec{\nabla}\frac{\delta F}{\delta\phi} - c\vec{\nabla}\frac{\delta F}{\delta c} + \eta_0\nabla^2\vec{v}, \tag{4}$$

where p is determined to satisfy the incompressibility condition. The viscosity η_0 is assumed, for simplicity, to be a constant independent of ϕ.

First we ignore the convective term of the third component c in Eq. (3). We choose $\sqrt{D/\gamma}$ as a characteristic length and $1/\gamma$ as a characteristic time of the problem. We derived the time-evolution equation of the scaled velocity of the droplet center as

$$\hat{m}\frac{d\hat{u}^\alpha}{d\hat{t}} = (-\tau_c + \hat{\tau})\hat{u}^\alpha - \hat{g}\hat{u}^\alpha\left|\hat{u}\right|^2, \tag{5}$$

Here $\hat{m}, \hat{\tau}$ and $\hat{g}$ are positive coefficients which depends only on the scaled radius $\hat{R} = R\sqrt{\gamma/D}$, and the dimensionless parameter τ_c is defined as

$$\tau_c = \frac{D^2\beta^3}{MA} = \frac{15\eta_0 D^2\beta^3}{2\sigma_1 A}. \tag{6}$$

All parameters except $\hat{R}$ are combined in the coefficient τ_c. Eq. (5) clearly indicates that changing the strength of the Marangoni effect induces a bifurcation from a motionless state to a propagating state.

The mechanism of mechanism of the droplet translational motion in our model for $A > 0$ and $\sigma_1 > 0$ is as follows. When a droplet is motionless, the concentration distribution of c around the droplet is isotropic. The concentration profile outside the droplet is a decreasing function of the distance from the center of mass. Let us suppose that the position of the droplet is shifted slightly. Then, the concentration of c decreases (increases) at the front (rear). Since the interfacial energy is an increasing function of c, the droplet tends to shift

further as shown schematically in Fig. 1. It is note that this argument can also be applied to the case that $A < 0$, $\sigma_1 < 0$, and $c(\vec{r}) \to c_\infty \neq 0$ in the limit $|\vec{r}| \to \infty$, which means that the third component is provided from the outside of the droplet and decomposed inside the droplet.

We estimated the effect of the convective term in Eq. (3), which has been ignored in the above treatment. We derive the correction from the convective term up to the first order of the perturbation expansion. The coefficient $\hat{\tau}$ is evaluated since this quantity is directly related to the drift instability threshold. In the limit $\hat{R} \to 0$, we obtain

$$\hat{\tau} = \frac{2\hat{R}^3}{15} P \tag{7}$$

When the convective term is not included, we have $P = 1$. On the other hand, the first order correction due to the convective term gives us $P = 31/56$. Since migration of droplet occurs for $\hat{\tau} \geq \tau_c$, this indicates that the stronger Marangoni effect is necessary when the convection of the third component exists.

We also carried out numerical integration of our model, and we observed the bifurcation from a motionless state to a stationary propagating state by changing the coefficient τ as shown in Fig. (2). As predicted theoretically, the composition profile of the third component becomes anisotropic around the droplet when the droplet undergoes self-propelled motion. We also confirmed that the convective term of the third dilute component suppresses the instability of a motionless state, which agrees with our analytical prediction in the limit $\hat{R} = R\sqrt{\gamma/D} \to 0$.

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[1] A. Shioi, T. Ban and Y. Morimune, Entropy 12, 2308 (2010).

[2] K. Nagai, Y. Sumino, H. Kitahata, and K. Yoshikawa, Phys. Rev. E 71, 065301(R) (2005).

[3] A. Ye. Rednikov and Y. S. Ryazantsev, J. Appl. Math. Mech. 53, 212 (1989).

[4] K. Kricher and A. Mikhailov Phys. Rev. Lett. 73 3165 (1994).

[5] T. Ohta, Physica D 151 61 (2001).

[6] S. Yabunaka, T. Ohta and N. Yoshinaga, J. Chem. Phys. 136 (2012): 074904.

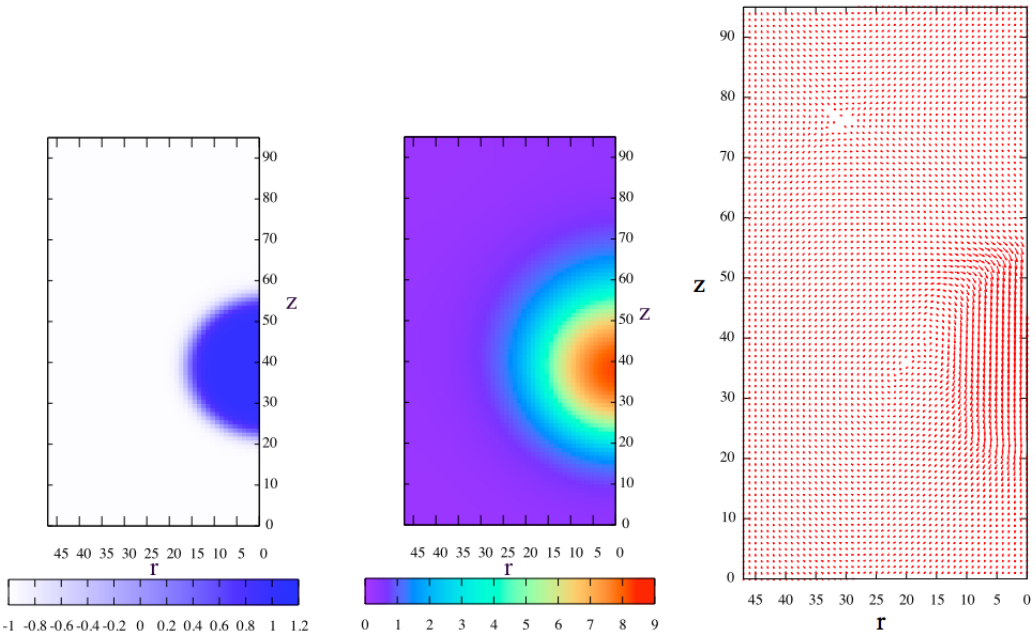

Figure 2: Stationary self-propelled motion of a droplet observed in a numerical simulation on a cylindrical cell (r, z). Displayed are (left) the composition ϕ, (center) the concentration of the third component c and (right) the velocity field $\vec{v}$. The horizontal axis corresponds to the coordinate r, and the vertical axis corresponds to the coordinate z.